PATENT SPECIFICATION

1,055,147

1,055,147

Date of Application and filing Complete Specification: September 25, 1963. *No. 37722/63*

Application made in United States of America (No. 229733) on October 10, 1962.

Complete Specification Published: January 18, 1967.

Index at Acceptance:—C2 B (B4B, B4H).

Int. Cl.:—C 07 d 1/14.

COMPLETE SPECIFICATION

NO DRAWINGS

Production of Ethylene Oxide

We, HALCON INTERNATIONAL, INC., of 2 Park Avenue, New York 16, New York, United States of America, a corporation organized and existing under the laws of the State of Delaware, United States of America

is the reaction of ethylene with molecular oxygen to form ethylene oxide, the desired product; the other is the reaction of ethylene with molecular oxygen to form carbon dioxide and water, which is an undesirable side

ERRATUM

SPECIFICATION NO. 1,055,147

Page 1, Heading Index at acceptance:- *for* "C2 B" *read* "C2 C"

THE PATENT OFFICE,
21st February, 1967

D 80597/17

ERRATA

SPECIFICATION NO. 1,055,147

Page 5, line 38, *for* "analoguos" *read* "analagous".

Page 5, line 59, *for* "0 001" *read* "0.001".

Page 5, line 108, *for* "0 01%" *read* "0.01%".

Page 5, line 118, *for* "0 4" *read* "0.4".

Page 6, line 6 and 7, *for* "selec vity" *read* "selectivity"

Page 6, line 12, *for* "0 0001" *read* "0.0001".

THE PATENT OFFICE,
30th March, 1967

D 83826/6

formation of carbon dioxide. At the same time maintaining optimum productivity.

In these ethylene oxide processes there are two main sources of yield loss. It is known in the art that two basic reactions occur, one

[Pr

suppresses the formation of both ethylene oxide and carbon dioxide.

It has now been found that, regardless of whether one is using air, oxygen, or a combination of air and oxygen as the oxidising

PATENT SPECIFICATION

1,055,147

1,055,147

Date of Application and filing Complete Specification: September 25, 1963. *No. 37722/63*

Application made in United States of America (No. 229733) on October 10, 1962.

Complete Specification Published: January 18, 1967.

Index at Acceptance:—C2 B (B4B, B4H).

Int. Cl.:—C 07 d 1/14.

COMPLETE SPECIFICATION

NO DRAWINGS

Production of Ethylene Oxide

We, HALCON INTERNATIONAL, INC., of 2 Park Avenue, New York 16, New York, United States of America, a corporation organized and existing under the laws of the State of Delaware, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to processes for the production of ethylene oxide by the partial oxidation of ethylene by means of gaseous oxygen in the presence of a silver containing catalyst.

The production of ethylene oxide by the partial oxidation of ethylene by means of gaseous oxygen in the presence of a silver containing catalyst is well known, and the suppression of excessive carbon dioxide formation in such a process has been recognized as a serious problem. Various methods have been proposed for attacking this problem, including the use of relatively large amounts of ethylene dichloride or the like materials, e.g., 300 p.p.m. based on the total gaseous mixture fed into the reactor. Experience with such methods, especially on rather large scales, has shown that the use of such amounts of ethylene dichloride or the like materials tends to kill or reduce the activity of the catalyst. It has been accepted by the art that as the concentration of ethylene oxide in outlet gases from the reactor increases, there necessarily is a decrease in reaction selectivity. The art is confronted by the problem of minimizing the excessive formation of carbon dioxide while at the same time maintaining optimum productivity.

In these ethylene oxide processes there are two main sources of yield loss. It is known in the art that two basic reactions occur, one is the reaction of ethylene with molecular oxygen to form ethylene oxide, the desired product; the other is the reaction of ethylene with molecular oxygen to form carbon dioxide and water, which is an undesirable side reaction. Selectivity is defined as the ratio of the ethylene which reacts to form oxide to the total ethylene which reacts, that is, S equals outlet oxide concentration minus inlet oxide concentration divided by inlet ethylene minus outlet ethylene concentration. Thus one source of yield loss is a poor selectivity when the ethylene is oxidized, giving mainly carbon dioxide and water. In a recycle process, in order to keep inert materials from building up in concentration, one must purge varying quantities of material from the system depending on the quantities of inerts fed. A certain portion of the feed ethylene is carried away in purging these impurities. Hence, another source of yield loss can be incomplete utilization of the ethylene or a poor overall C_2H_4 conversion. The overall conversion is related to the conversion per pass in the reactor and the amount of material recycled as well as the concentration of ethylene in the purge from the system. In any case whether one is using air or oxygen as the oxidizing medium, a purge must be taken from the system in order to keep the inert material such as argon, nitrogen, CH_4 or C_2H_6 from building up in the system. One must also remove carbon dioxide from the system, either by scrubbing the recycle stream to remove it, or by taking a sufficiently large purge stream to keep its concentration in an acceptable range, since carbon dioxide acts as an inhibitor in the system and suppresses the reaction of ethylene to form both ethylene oxide and carbon dioxide.

It has now been found that, regardless of whether one is using air, oxygen, or a combination of air and oxygen as the oxidising

[Pr———.]

medium, there are certain critical reaction conditions required depending on what process one is practising, that is, there is a critical inhibitor concentration for given concentrations of carbon dioxide, ethane, methane, ethylene, and oxygen in the system. The critical condition is also a function of the particular reactor temperature, pressure and contact time, and tube diameter. This control of inhibitor, carbon dioxide, and ethane and/or methane concentration results in marked improvement in the process yield and reactor productivity. For this invention, molecular oxygen from some source is reacted with ethylene over a silver catalyst, and the invention can be practised using catalyst in either carbon steel tubes or stainless steel tubes, such as, for example 304, 316, 405 or 410 stainless steels. Materials of construction have no effect on the results. To practise this invention one must keep the carbon dioxide concentration to the inlet of the reactor below 10%, and preferably in the range of 3 to 8%, either by scrubbing out carbon dioxide from the recycle gases or by taking a sufficiently large purge from the system to maintain the carbon dioxide level in this range.

According to the invention a process for producing ethylene oxide by the partial oxidation of ethylene by means of gaseous oxygen in the presence of a silver containing catalyst, comprises operating with a concentration of oxygen in the range of 1 to 10%, a concentration of ethylene in the range of 1 to 30%, a concentration of carbon dioxide in the range of 0.1 to 10%, a concentration of halogenated inhibitor in the range of 0.0001 to 100 p.p.m., a concentration of methane and/or ethane in the range of 0.001 to 40%, a reactor temperature in the range of 150° to 400°C., a reactor pressure in the range of 50 to 650 p.s.i.g. a flow rate in the range of 50 to 1500 standard cubic feet per hour (hereinafter referred to as "S.C.F.H.") per elongated zone with the zone average diameter in the range of 0.5 to 2.0 ins. and the zone length in the range of 5 to 40 ft., the particular combination of conditions being selected and controlled to provide a selectivity in the range of 60% to 80% and an ethylene oxide concentration in the exit gas in the range of 0.3 to 5.0%.

In the process of the invention, set forth in the preceding paragraph:

(a) the feed may contain 5% ethylene, 0.01 to 0.1% ethane and/or methane, 5 to 7% carbon dioxide and 0.01 to 0.1 p.p.m. of ethylene dichloride inhibitor, the pressure may be 300 p.s.i.g., the selectivity may be 70% to 73% and the ethylene oxide concentration in the exit gas may be 0.4 to 1.3%;

(b) the feed may contain 0.05 p.p.m. of inhibitor, the temperature may be 250°C., the selectivity may be 73% and the ethylene oxide concentration in the exit gas may be about 1%;

(c) the feed may contain 10% of oxygen, 30% of ethylene, 40% of ethane and/or methane or both, 0.3 to 40 p.p.m. of ethylene dichloride and 5% of carbon dioxide, the flow rate may be 550 S.C.F.H., the selectivity may be 71% and the ethylene oxide concentration in the exit gas may be 1.0 to 2.0%;

(d) the feed may contain about 0.1 p.p.m. of inhibitor, the temperature may be 260°C. the selectivity may be 70% and the ethylene oxide concentration in the exit gas may be 1.114%;

(e) the feed may contain 0.04 p.p.m. of inhibitor and 260°C. at a selectivity of 70% and the ethylene oxide concentration in the exit gas may be 0.963%;

(f) the feed may contain 0.125 p.p.m. of inhibitor and a flow rate of 850 at 260°C. at a selectivity of 70% and the ethylene oxide concentration in the exit gas may be 1.161%;

(g) the feed may contain about 0.005 p.p.m. of inhibitor and 0.01% ethane, the pressure may be 150 p.s.i.g. the selectivity may be 70% and the ethylene oxide concentration in the exit gas may be 0.755%;

(h) the feed may contain 0.289 p.p.m. of inhibitor and 0.01% ethane, the pressure may be 500 p.s.i.g. the selectivity may be 70% and the ethylene oxide concentration in the exit gas may be 1.239%;

(i) the feed may contain 20% ethylene, 8% ethane and/or methane, 5% carbon dioxide and 0.03 to 45.0 p.p.m. of ethylene dichloride inhibitor, the pressure may be 300 p.s.i.g., the selectivity may be 70 to 73% and the ethylene oxide concentration in the exit gas may be 0.4 to 3.3%;

(j) the feed may contain 0.0035 p.p.m. of inhibitor and 6% oxygen, the flow rate may be 550 S.C.F.H. (standard cubic feet per hour), the temperature may be 250°C., the selectivity may be 70% and the ethylene oxide concentration in the exit gas may be 2.62%;

(k) the feed may contain 0.0411 p.p.m. of inhibitor and 10% oxygen, the flow rate may be 550 S.C.F.H., the temperature may be 250°C., the selectivity may be 70% and the ethylene oxide concentration in the exit gas may be 3.15%;

(l) the feed may contain 30% ethylene, 40% ethane and/or methane, 10% oxygen and 0.03 to 45 p.p.m. of ethylene dichloride inhibitor, the pressure may be 300 p.s.i.g., the selectivity may be 70 to 73% and the ethylene oxide concentration in the exit gas may be 0.4 to 3.3%;

(m) the feed may contain 1 to 6% of ethy-

lene, 4 to 7% of oxygen, 0.001 to 1% of ethane and/or methane, 0.0001 to 0.2 p.p.m. of ethylene dichloride inhibitor and 1 to 9% of carbon dioxide, and the selectivity may be 60 to 80%;

(n) the feed may contain 1 to 3% of ethylene, the selectivity may be 60 to 68%, and the ethylene oxide concentration in the exit gas may be 0.5 to 1.2%;

(o) the feed may contain 4 to 6% of ethylene and 1 to 8% of carbon dioxide, the selectivity may be 68 to 74% and the ethylene oxide concentration in the exit gas may be 0.5 to 2.0%;

(p) the feed may contain 1 to 3% of ethylene, 4 to 7% of oxygen, 0.001 to 0.1% of ethane and/or methane and 1 to 9% of carbon dioxide, the selectivity may be 60 to 68% and the ethylene oxide concentration in the exit gas may be 0.5 to 1.2%;

(q) the feed may contain 10 to 30% of ethylene, 4 to 10% of oxygen, 0.001 to 40% of ethane and/or methane, 0.01 to 100 p.p.m. of ethylene dichloride inhibitor and 1 to 8% of carbon dioxide, the selectivity may be 65 to 80% and the ethylene oxide concentration in the exit gas may be 0.5 to 5.0%.

In order to indicate still more fully the nature of the present invention, the following examples of typical procedures are set forth in which, as in the foregoing description and in the claims, parts and percents mean parts and percents by voulme, respectively, unless otherwise indicated, it being understood that these examples are presented as illustrative only and they are not intended to limit the scope of the invention.

EXAMPLE 1

Particulate silver catalyst material is placed in 1" diameter vertical steel tubes, (carbon or stainless steel) to a catalyst bed height of 25 feet. A gaseous reaction mixture is prepared containing by volume 5% ethylene, 6% oxygen, 0.1% ethane, 7% carbon dioxide, 0.05 p.p.m. ethylene dichloride as inhibitor and the rest nitrogen and other inert gases. This gaseous reaction mixture is passed at a flow rate of 650 standard cubic feet per hour per tube through the catalyst bed at 250°C. and 300 p.s.i.g. The ethylene is oxidized to ethylene oxide with a 73% selectivity at a conversion of 27%. The exit gases contain about 1% ethylene oxide.

The product mixture from this reaction is scrubbed with water to remove product ethylene oxide. A portion of gas is purged in order to prevent a build up of inerts in the system. A portion of the gases may be scrubbed with potassium carbonate solution to reduce the carbon dioxide concentration and is then recycled to the initial reaction. Sufficient air or oxygen, ethylene, ethane or methane and ethylene dichloride are added to this recycle mixture to provide the charge mixture to the reaction as above described.

In analogous runs, the carbon dioxide concentration at the reaction inlet is built up to and then reduced from 20% to 10%; the ethylene oxide production is increased by 17%, with no loss in efficiency or selectivity. If the carbon dioxide in the reactor inlet is reduced from 20% to 5% the ethylene oxide production is increased by about 38%. Hence, it is beneficial to keep the carbon dioxide concentration at a low level namely in the range of 0.1 to 10%.

In other analogous runs at 260°C., a flow rate of about 550, an ethane and/or methane content of 0.01%, at a selectivity of 70%, 0.0997 p.p.m. of the inhibitor gives exit gas containing 1.114% of ethylene oxide. Dropping the inhibitor to 0.04 p.p.m. gives an exit gas containing 0.963% of ethylene oxide. Raising the flow rate to about 850, and using 0.125 p.p.m. of inhibitor gives an exit gas containing 1.161% of ethylene oxide. In other runs at 250°C., with 0.01% ethane or methane at a pressure of 150 p.s.i.g. and using 0.000493 p.p.m. of inhibitor, the exit gas contains 0.755% of ethylene oxide. Raising the pressure to 500 p.s.i.g. and using 0.289 p.p.m. of inhibitor gives an exit gas containing 1.239% of ethylene oxide.

From these data, it is apparent that the invention enables one to operate at good efficiency with high productivity.

EXAMPLE 2

In this embodiment of the invention, pure oxygen, (i.e., 85-99.9%) is used as oxidant. The same catalyst, reactor and flow rate is used as described in Example 1. A gaseous inlet mixture to the reactor is prepared containing about 20% ethylene, 8% oxygen, 8% ethane or methane, 5% carbon dioxide and about 30 p.p.m. ethylene dichloride. At a reaction temperature of 235°C. and 300 p.s.i.g. about 14% of the ethylene reacts upon passage through the catalyst bed with a 73% selectivity to ethylene oxide. The exit gases contain about 2% ethylene oxide. The reactor effluent is scrubbed with water to remove ethylene oxide and then is scrubbed with aqueous potassium carbonate to reduce the carbon dioxide content of these gases. The thusly scrubbed gases are recycled to the reaction and sufficient ethylene, oxygen, ethane or methane and ethylene dichloride and sometimes air or nitrogen are added to give the above specified reaction inlet gas mixture.

By way of contrast, where Example 2 is duplicated and the carbon dioxide level is not reduced by scrubbing but is allowed to rise to about 20% carbon dioxide in the inlet reaction mixture, there is only about a 6% conversion of ethylene per pass, and the reactor exit gases contain only about 1% ethylene oxide.

If one increases the ethane or methane concentration in the feed to the reactor from 0% to 1% and at the same time raises the inhibitor concentrations from 0.02 p.p.m. to 0.10 p.p.m. then one achieves a 30% increase in productivity, at the same selectivity. Alternatively, one can increase the selectivity from 65% to 72%, at the same ethylene oxide production, by increasing inhibitor concentration and raising ethane or methane. Hence, it is desirable for one to control the amount of ethane or methane in the ethylene feed to the system, either by adding supplementary methane or ethane, or by careful control in the ethylene plant to give a specified composition of ethylene. Changes in methane or ethane concentrations in the reactor inlet are compensated for by changes in inhibitor addition rate.

Following the Example 2 conditions using 20% ethylene, 30 p.p.m. of inhibitor and a 70% selectivity the reactor outlet gas contains 2% ethylene oxide. This is a 2-fold increase over the content where one uses 5% ethylene, 0.04 p.p.m. of inhibitor and a 70% selectivity. By increasing the inhibitor concentration at the same time as one increases the ethylene or oxygen concentration or decreases the carbon dioxide concentration, one can maintain the reactor efficiency at the same level while increasing the productivity by a factor of two (2).

In analogous runs at 250°C., a flow rate of about 550, an ethane content of 0.01%, a selectivity of 70, 6% oxygen and 5% carbon dioxide, 0.00334 p.p.m. of inhibitor gives an exit gas containing 2.62% of ethylene oxide. Raising the oxygen to 10% and the inhibitor to 0.0411 p.p.m. gives an exit gas containing 3.15% of ethylene oxide; however, if the ethane and/or methane content is raised to 20% and the inhibitor to 42.6 p.p.m. the exit gas contains 0.800% of ethylene oxide.

In another run using 30% ethylene, 40% ethane and/or methane, 40 p.p.m. of ethylene dichloride at 300 p.s.i.g. at a selectivity of 73% the ethylene oxide concentration in the exit gas is 3.0%.

The invention enables one to operate with good efficiency (in the range of 60 to 80 selectivity) and high productivity (in the range of 0.3 to 5.0% of ethylene oxide in the exit gas).

In practising the ethylene oxide process where one uses relatively pure oxygen as a feed-stock instead of air, then one can operate at economically higher inlet ethylene concentrations, since, less purge must be taken from the system, due to the fact that a smaller amount of inert materials is being fed with the oxygen stream; i.e. 0.1-10% inerts with pure oxygen versus 79% inerts in air. Hence, the amount of purge gas relative to fresh ethylene fed in the air process is approximately seven (7) pounds of purge gas per pound of fresh ethylene fed, whereas, if one is feeding a stream containing 95% oxygen instead of 20% oxygen, then the amount of purge is only about 0.1 pounds of ethylene per pound of ethylene fed. Hence, in order to get the same concentration in the purge from the system if one operates with oxygen, the ethylene concentration in the purge gas can be seventy times greater than in the air case for the same overall ethylene conversion. Since, the overall ethylene conversion is given by the following formula:

$$Ko = 1 - C_2H_{4p}P/F$$

where C_2H_{4p} is mol fraction of C_2H_4 in purge gas from the system, P/F is pounds of purge gas from the system divided by pounds of fresh ethylene fed. The value of P/F is primarily a function of the amount of inerts fed to the system. Hence, if one feeds air with 21% O_2 and 79% N_2 then P/F is about 7.0. If one feeds a stream with 40% O_2 and 60% N_2 then P/F is about 2.2. If one uses 60% O_2 and 40% N_2 then P/F is about 1.0, or if one uses 95% O_2 and 5% N_2 then P/F is about 0.1. These values are true regardless of the number of reaction systems in series or parallel. For example, one can have a main reaction system with recycle and the purge from it being fed to a purge reaction system with or without recycle. Also the gases purged from the purge reaction systems could be fed to a secondary purge reaction system with or without recycle. All the concepts presented in this invention apply to all of these types of systems.

As the purge to feed ratio decreases due to the fact that fewer inerts are being added to the system, one can go to higher ethylene concentrations without taking an overall yield loss due to poor ethylene utilization. This invention provides substantial improvements in the ethylene oxide production yield by control of the inlet concentrations of lower saturated hydrocarbon (methane and ethane) and carbon dioxide and inhibitor in the ethylene oxide system. Also, supplementary addition of inerts, such as nitrogen or argon enables one to achieve better control of the reaction system. In order to get optimum performance all of these variables must be carefully regulated and controlled. The optimum combination of these variables is readily determined in accordance with the invention for any particular catalyst, reactor configuration, the pressure of operation, the gas flow rate to the catalyst bed, and the tube geometry, as well as the system operating pressure. In all cases the concentration at the reactor inlet is kept below 10% in order to prevent getting into the range where an explosion or detonation can occur. This is independent of the amount of hydrocarbon or type of hydrocarbon present in the system.

In order to get good efficiency in the process of the invention one must always add

inhibitor, within the range 0.0001 to 100 p.p.m.

The catalyst employed in the foregoing examples is regarded as particularly effective and desirable. It has a long active life. Other silver containing catalysts may be used however, such as those known to the art.

Air or relatively pure oxygen may be used to provide the oxygen required. The reaction is conducted at temperatures in the range of 150°C. to 400°C., preferably about 225° to 300°C. The reaction is conducted under a pressure of from 50 to 650 p.s.i.g. The reaction mixture, temperature, catalyst, and contact time or space velocity of the gaseous mixtures are interrelated and suitable combinations thereof are selected to give the desired optimum output or selectivity and concentration of ethylene oxide in the exit gas.

The process may be carried out in a manner wherein the reacted gases are scrubbed, e.g., with water or the like, to remove ethylene oxide product, and then are recycled to the reactor together with additional ethylene, C_2H_6, CH_4, A or N_2, and air and ethylene dichloride to maintain the desired reaction mixture concentration with purging of part of the scrubbed gases to maintain the desired volume and pressure conditions in the system. Care should be taken in such an operation to avoid the presence of excessive amounts of ethylene dichloride in the reaction zone to avoid deleterious effects on the catalyst.

Although ethylene dichloride is indicated as a suitable halogenated inhibitor material, other halogen or halogen containing materials having analoguos vapour pressure to ethylene dichloride or being gaseous or low boiling or having a boiling point up to about 300 or 350°C. may be used, especially where all of the advantages of ethylene dichloride are not necessarily required. Typical examples of such other materials are other halogenated hydrocarbons, halogens, hydrohalides, ammonium or the like halides.

WHAT WE CLAIM IS:—

1. A process for producing ethylene oxide by the partial oxidation of ethylene by means of gaseous oxygen in the presence of a silver containing catalyst, which comprises operating with a concentration of oxygen in the range of 1 to 10%, a concentration of ethylene in the range of 1 to 30%, a concentration of carbon dioxide in the range of 0.1 to 10%, a concentration of halogenated inhibitor in the range of 0.0001 to 100 p.p.m., a concentration of methane and/or ethane in the range of 0.001 to 40%, a reactor temperature in the range of 150° to 400°C., a reactor pressure in the range of 50 to 650 p.s.i.g., a flow rate in the range of 50 to 1500 standard cubic feet per hour per elongated zone with the zone average diameter in the range of 0.5 to 2.0 ins. and the zone length in the range of 5 to 40 ft., the particular combination of conditions being controlled to provide a selectivity in the range of 60% to 80% and an ethylene oxide concentration in the exit gas in the range of 0.3 to 5.0%.

2. A process as claimed in claim 1, in which the feed contains 5% ethylene, 0.01 to 0.1% ethane and/or methane, 5 to 7% carbon dioxide and 0.01 to 0.1 p.p.m. of ethylene dichloride inhibitor, the reactor pressure is 300 p.s.i.g., the selectivity is 70% to 73% and the ethylene oxide concentration in the exit gas is 0.4 to 1.3%.

3. A process as claimed in claim 1, in which the feed contains 0.05 p.p.m. of inhibitor, the reactor temperature is 250°C., the selectivity is 73% and the ethylene oxide concentration in the exit gas is about 1%.

4. A process as claimed in claim 1, in which the feed contains about 0.1 p.p.m. of inhibitor, the reactor temperature is 260°C., the selectivity is 70% and the ethylene oxide concentration in the exit gas is 1.114%.

5. A process as claimed in claim 1, in which the feed contains 0.04 p.p.m. of inhibitor, the reactor temperature is 260°C., the selectivity is 70% and the ethylene oxide concentration in the exit gas is 0.963%.

6. A process as claimed in claim 1, in which the feed contains 0.125 p.p.m. of inhibitor, the flow rate is 850, the reactor temperature is 260°C., the selectivity is 70% and the ethylene oxide concentration in the exit gas is 1.161%.

7. A process as claimed in claim 1, in which the feed contains about 0.005 p.p.m. of inhibitor and 0.01% ethane, the pressure is 150 p.s.i.g., the selectivity is 70% and the ethylene oxide concentration in the exit gas is 0.755%.

8. A process as claimed in claim 1, in which the feed contains 0.289 p.p.m. of inhibitor and 0.01% ethane, the pressure is 500 p.s.i.g., the selectivity is 70% and the ethylene oxide concentration in the exit gas is 1.239%.

9. A process as claimed in claim 1, in which the feed contains 20% ethylene, 8% ethane and/or methane, 5% carbon dioxide, 0.03 to 45.0 p.p.m. of ethylene dichloride inhibitor, the pressure is 300 p.s.i.g., the selectivity is 70% to 73% and the ethylene oxide concentration in the exit gas is 0.4 to 3.3%.

10. A process as claimed in claim 1, in which the feed contains 0.0035 p.p.m. of inhibitor and 6% oxygen, the flow rate is 550 S.C.F.H., the reactor temperature is 250°C., the selectivity is 70% and the ethylene oxide concentration in the exit gas is 2.62%.

11. A process as claimed in claim 1, in which the feed contains 0.0411 p.p.m. of inhibitor and 10% of oxygen, the flow rate is 550 S.C.F.H., the reactor temperature is 250°C., the selectivity is 70% and the ethylene oxide concentration in the exit gas is

3.15%.

12. A process as claimed in claim 1, in which the feed contains 30% ethylene, 40% ethane and/or methane, 10% oxygen and 0.03 to 45 p.p.m. of ethylene dichloride inhibitor, the pressure is 300 p.s.i.g., the selectivity is 70 to 73% and the ethylene oxide concentration in the exit gas is 0.4 to 3.3%.

13. A process as claimed in claim 1, in which the feed contains 1 to 6% of ethylene, 4 to 7% of oxygen, 0.001 to 1% of ethane and/or methane, 0.0001 to 0.2 p.p.m. of ethylene dichloride inhibitor, and 1 to 9% of carbon dioxide and the selectivity is 60 to 80%.

14. A process as claimed in claim 1, in which the feed contains 1 to 3% of ethylene, the selectivity is 60 to 68%, and the ethylene oxide concentration in the exit gas is 0.5 to 1.2%.

15. A process as claimed in claim 1, in which the feed contains 4 to 6% of ethylene and 1 to 8% of carbon dioxide, the selectivity is 68 to 74% and the ethylene oxide concentration in the exit gas is 0.5 to 2.0%.

16. A process as claimed in claim 1, wherein the feed contains 1 to 3% of ethylene, 4 to 7% of oxygen, 0.001 to 0.1% of ethane and/or methane and 1 to 9% of carbon dioxide, the selectivity is 60 to 68% and the ethylene oxide concentration in the exit gas is 0.5 to 1.2%.

17. A process as claimed in claim 1, in which the feed contains 10 to 30% of ethylene, 4 to 10% of oxygen, 0.001 to 40% of ethane and/or methane, 0.01 to 100 p.p.m. of ethylene dichloride inhibitor and 1 to 8% of carbon dioxide, the selectivity is 65 to 80% and the ethylene oxide concentration in the exit gas is 0.5 to 5.0%.

18. A process for producing ethylene oxide according to claim 1 substantially as hereinbefore described and exemplified.

19. Ethylene oxide when produced by the process claimed in any of the preceding claims.

MATHYS & SQUIRE,
Chartered Patent Agents,
Staple House, 51/52, Chancery Lane,
London, W.C.2.
Agents for the Applicants.

Berwick-upon-Tweed: Printed for Her Majesty's Stationery Office by The Tweeddale Press Ltd.—1966
Published at the Patent Office 25 Southampton Buildings, London. W.C.2 from which copies may be obtained.